



UNITED STATES DEPARTMENT OF COMMERCE
National Oceanic and Atmospheric Administration
NATIONAL MARINE FISHERIES SERVICE

Northwest and Alaska Fisheries Center
Environmental Conservation Division
2725 Montlake Boulevard East
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August 7, 1984

F/NWC6:DCM

Dr. Gary O'Neal
Chief, Environmental Services Division
U.S. Environmental Protection Agency
M/S 337
1200 Sixth Avenue
Seattle, WA 98101

Dear Gary:

Dan Petke asked us to submit a summary report on our chemical and biological data from Eagle Harbor. We are pleased to provide this information herewith.

Samples of sediments and English sole were obtained from Eagle Harbor on December 8, 1983 and April 5, 1984. The sampling of Eagle Harbor was undertaken in connection with our on going studies of toxic chemicals and alterations in the health of Puget Sound marine life. The Eagle Harbor study was conducted in concert with complimentary work undertaken by EPA/DOE.

Aromatic Hydrocarbons in Sediment

Concentrations of total hydrocarbons (dry weight) in Eagle Harbor sediments are given in Figure 1. Detailed analyses of these hydrocarbons are presented in Tables I and II. In addition a variety of nitrogen-containing aromatic compounds, including the liver carcinogen carbazole, have been identified. Chlorinated organic compounds, including PCB's were found only in trace amounts. It is apparent that high concentrations of aromatic hydrocarbons, resembling those of creosote, are present in the sediments of a major portion of Eagle Harbor. In fact, a number of the sites examined contained hydrocarbon concentrations far exceeding those in Seattle's highly polluted Duwamish River.

Metals in Sediment

The concentrations of metals in sediments from Eagle Harbor and from a reference area (President Point) were generally similar. Details are available upon request.

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Aromatic Hydrocarbons in English Sole

Concentrations of aromatic hydrocarbons in the stomach contents, liver and muscle of English sole are given in Figure 2. Relatively high concentrations of aromatic hydrocarbons were found, compared to the stomach contents (comprising mostly invertebrates) from fish obtained from a reference area (Port Jefferson). These data indicate that sediment-dwelling organisms that flatfish feed on appear to be a significant source of chemical exposure. Although the concentration of hydrocarbons in the sole liver is less than 1000 ppb, this value is high for exposed fish because the liver extensively converts hydrocarbons to other products. It is noteworthy that the broad scan chemical analyses of edible muscle from English sole from heavily polluted areas of Eagle Harbor failed to reveal evidence of more than trace amounts of toxic chemicals. Such trace amounts are characteristic of fish tissue from essentially non-polluted areas of Puget Sound. This finding is consistent with data from a number of studies from our laboratories.

Metabolites of Aromatic Compounds in Bile of English Sole

Two samplings indicated that metabolites structurally similar to benzo(a)pyrene were present in substantially higher concentrations in the bile of English sole from Eagle Harbor than in the bile of fish from a reference area (President Point). These findings indicate that the English sole were exposed to aromatic hydrocarbons and converted them to potentially carcinogenic metabolites.

Short-term Bioassays of Eagle Harbor Sediment

Results of a variety of bioassays employing diverse test organisms indicated that Eagle Harbor sediments with the highest concentrations of hydrocarbons were acutely toxic. The evidence is summarized in Table III.

Diseases in English Sole

Gross and histopathologic examinations of English sole from Eagle Harbor were performed and the prevalences of liver tumors and other abnormalities of the liver are given in Figure IV. The evidence indicates that a large portion of the English sole population is afflicted with liver tumors (20 to 30%) and degenerative diseases of the liver (~90%)--diseases which have been linked to toxic chemical pollution in other areas of Puget Sound.

Summary

The findings from our laboratories indicate that a major portion of Eagle Harbor is severely contaminated with aromatic hydrocarbons of apparent creosote origin. These hydrocarbons are taken up by English

sole and metabolized to potentially toxic substances, some of which have been linked to neoplastic diseases in laboratory studies. The essentially base-line concentration of the actively metabolized aromatic hydrocarbons in the fish muscle was not unexpected. It should not be construed, however, that the muscle of bottom-dwelling fish from other polluted areas will necessarily also have comparably low concentrations of other potential toxic chemicals.

Commensurate with the high degree of hydrocarbon pollution in Eagle Harbor are indications of serious acute and chronic biological effects. It is especially noteworthy that each of the six bioassays routinely used in our laboratories showed the sediments to be extremely toxic. In addition, the high prevalences of liver tumors and other liver abnormalities in fish obtained from several locations in Eagle Harbor are clearly long-term biological effects that are linked to the hydrocarbon exposure. In our experience, the high concentrations of toxic organic chemicals (i.e., aromatic hydrocarbons) in Eagle Harbor sediments and serious associated biological effects are unparalleled elsewhere in Puget Sound.

Sincerely,



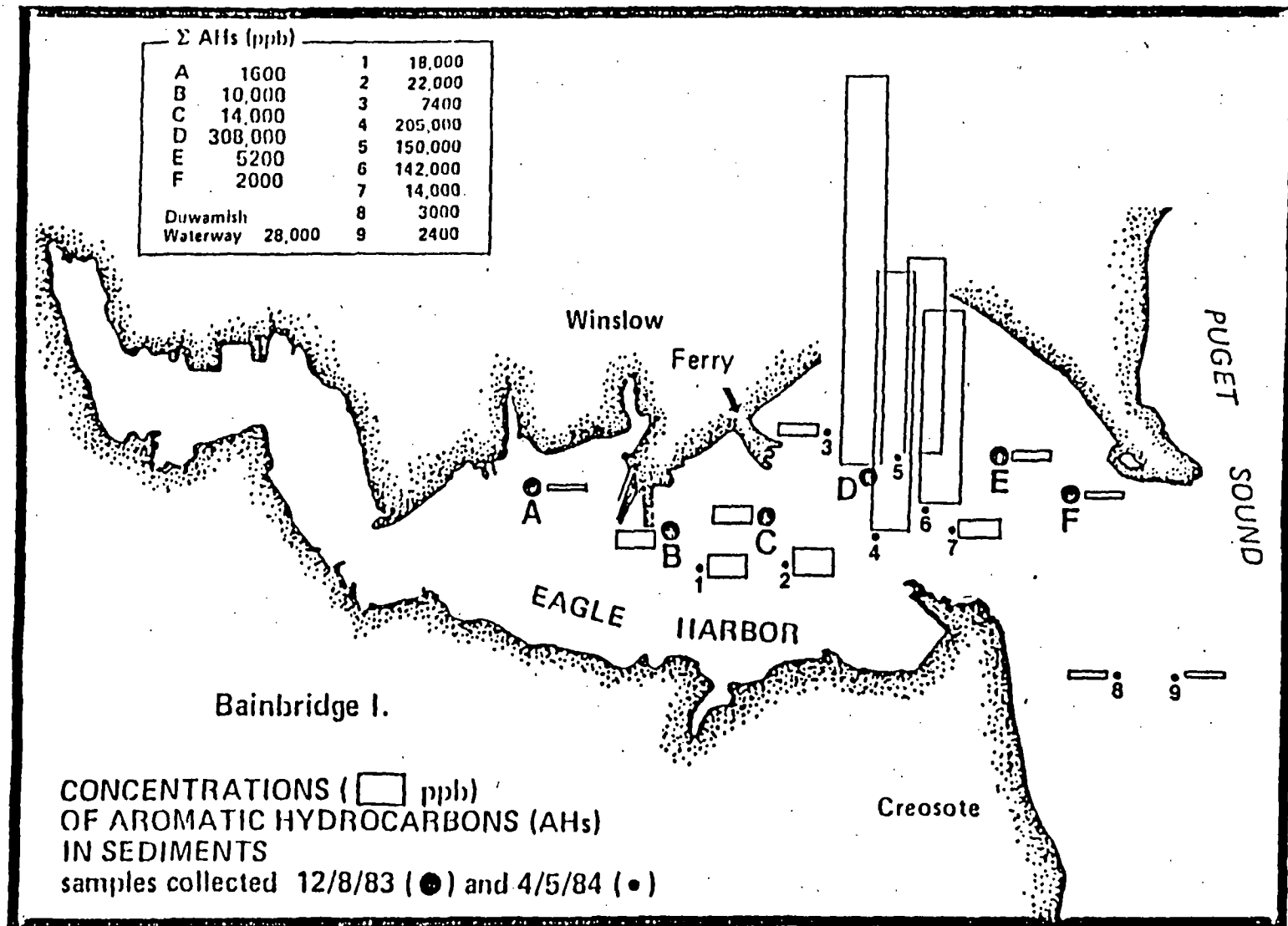
Donald E. Malins, PhD, DSc.
Division Director

cc: D. Petke, EPA
D. Ancona, GC

REFERENCES

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- MALINS, D.C., B.B. MCCAIN, D.W. BROWN, S-L. CHAN, M.S. MYERS, J.T. LANDAHL, P.G. PROHASKA, A.J. FRIEDMAN, L.D. RHODES, D.G. BURROWS, W.D. GRONLUND and H.O. HODGINS (1984). Chemical pollutants in sediments and diseases in bottom-dwelling fish in Puget Sound, Washington. *Environ. Sci. Technol.* (In press).

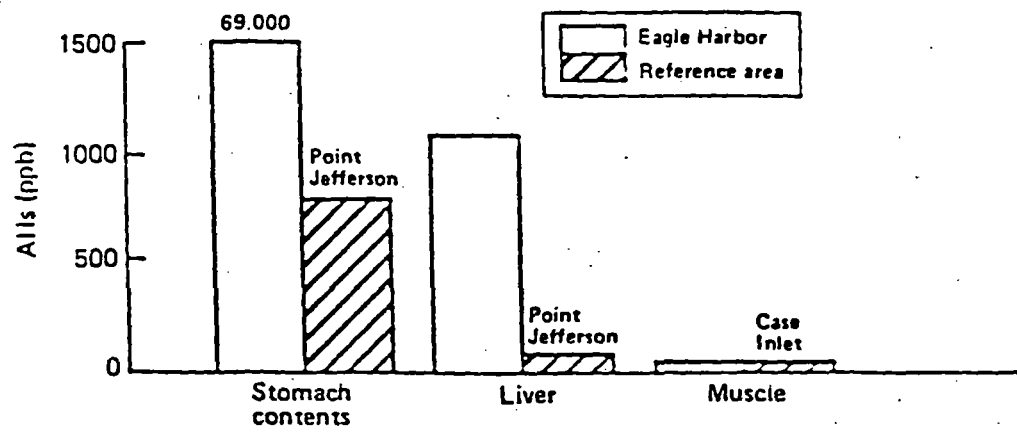
FIGURE 1



Reference to analytical procedures: Malins et al. 1984.

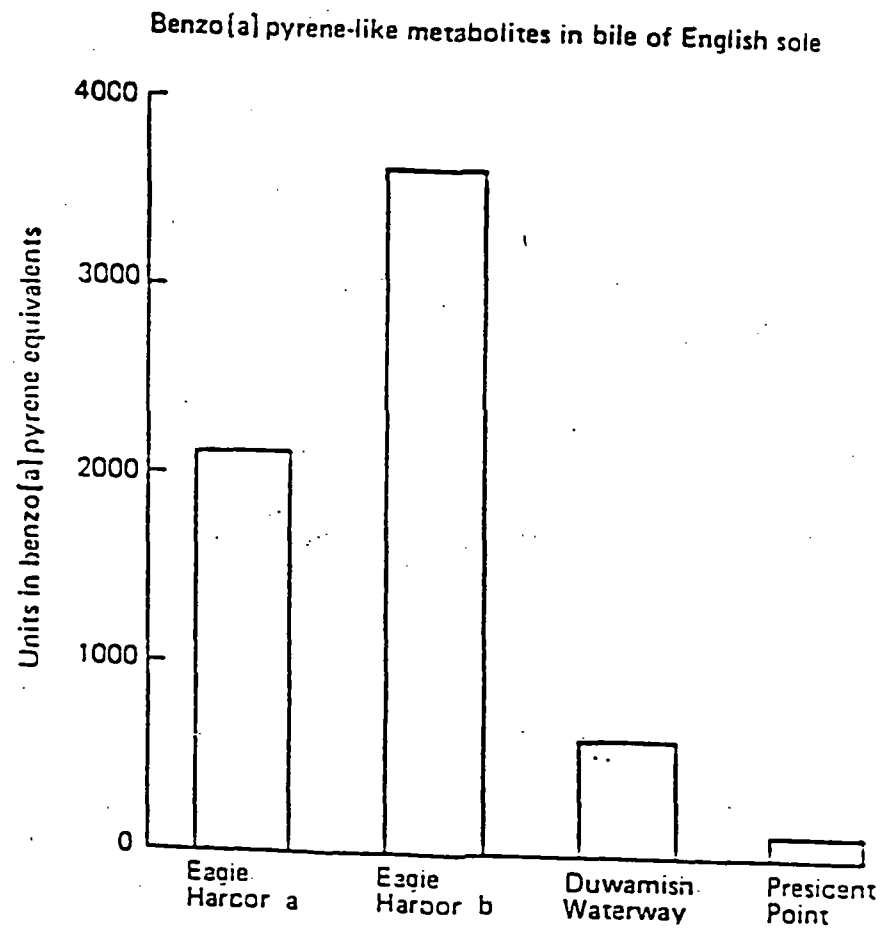
FIGURE 2

Concentrations of Aromatic Hydrocarbons in English sole



Reference to analytical procedures: Malins et al. 1984.

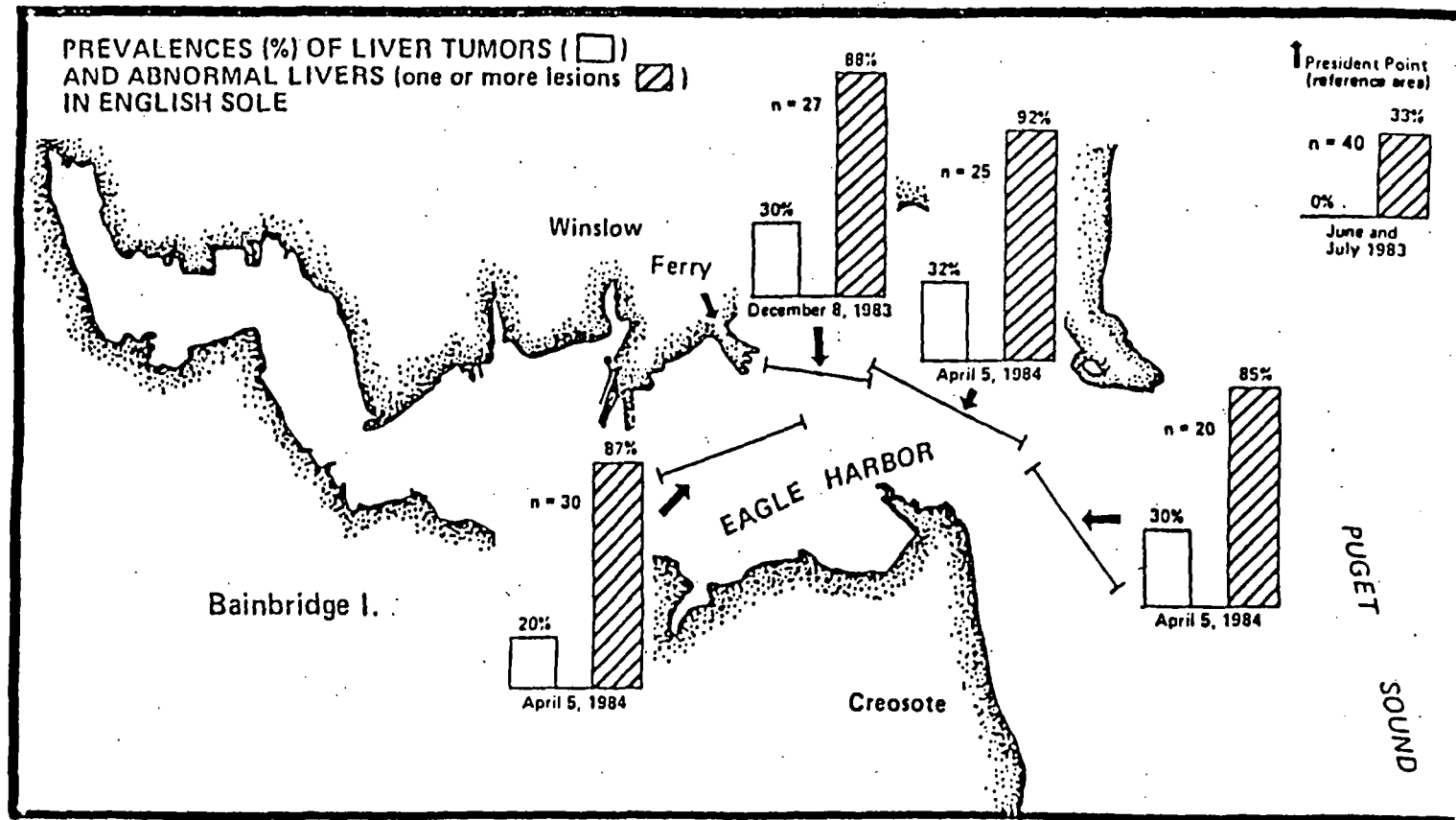
FIGURE 3



Footnotes: a. Sampled 12/8/83
b. Sampled 5/31/84

Reference to analytical procedures: Krahn et al. 1984.

FIGURE 4



Reference to analytical procedures: Malins et al. 1984.

Table 1.

a,b,c,d

Concentrations of aromatic compounds in sediment samples, ng/g (ppb) dry weight.

Site-Station:	EGH-A	EGH-B	EGH-C	EGH-D	EGH-E	EGH-F	PPT	SPIKE
1-Propyl Benzene	< 2.8	< 15	< 11	< 20	< 5.3	< 2.5	< 4.2	< 2.8
n-Propyl Benzene	< 2.8	< 15	< 11	< 20	< 5.5	< 2.6	< 4.4	< 2.8
Indan	2.9	< 14	< 10	290	5.4	< 2.4	< 4.2	< 2.6
1,2,3,4-Tetraethylbenzene	< 2.5	< 14	< 9.7	26	< 4.8	< 2.3	< 4.3	< 2.5
Naphthalene	98	220	310	8800	270	44	8.9	< 2.0
Benzothiophene	5.6	< 16	19	470	14	< 2.7	< 5.1	< 3.0
2-Methyl Naphthalene	28	59	86	5400	73	8.2	< 3.5	< 2.0
1-Methyl Naphthalene	16	30	43	5500	37	4.4	< 3.5	< 1.9
Biphenyl	10	20	38	590	33	2.4	< 1.7	< 2.0
2,6-Dimethyl Naphthalene	9.9	29	39	2200	36	< 1.8	< 1.8	< 2.1
Acenaphthene	34	53	84	22000	98	5.3	< 1.9	< 2.1
2,3,5-Trimethyl Naphthalene	< 2.0	< 11	< 7.4	1800	< 3.8	2.7	< 1.9	< 2.1
Fluorene	45	110	170	26000	140	19	34	< 2.2
Dibenzothiophene	28	92	120	9500	38	12	< 2.1	< 2.2
Phenanthrene	130	600	700	76000	470	73	150	< 1.7
Anthracene	65	350	570	25000	220	100	150	< 1.7
1-Methyl Phenanthrene	9.5	71	51	3400	36	7.7	< 1.5	< 1.6
3,6-Dimethyl Phenanthrene	3	27	30	1000	< 3.1	< 1.5	7.3	< 1.7
Fluoranthene	180	1200	1400	59000	770	94	220	< 1.6
Pyrene	240	1600	1800	32000	800	140	93	< 1.6
Benz[a]anthracene	99	820	1100	9300	370	200	71	< 2.6
Chrysene	180	1400	2200	11000	670	450	140	< 3.1
Benzofluoranthene	100	900	1200	2400	260	160	100	< 2.0
Benzofluoranthene	85	760	1200	2300	300	210	43	3.6
Benzofluoranthene	98	740	940	2300	240	210	41	< 2.6
Perylene	18	190	270	530	64	45	17	< 2.1
Indenopyrene	42	400	520	480	110	100	30	< 3.0
Bibenz[a,h]anthracene	11	120	190	300	37	30	8.3	< 3.0
Benzofluoranthene	37	330	470	640	100	84	23	5.2
Sample Weight (g)	20.00	20.00	20.00	20.06	20.02	20.01	20.03	20.01
% Dry Weight	78.07	51.72	51.88	63.34	74.92	79.65	78.29	67.24
Recovery of DB Naphthalene	93%	79%	61%	100% e	85%	96%	72%	96%
Recovery of D10 Acenaphthene	83%	72%	56%	100% e	75%	84%	79%	81%
Recovery of D12 Perylene	71%	64%	52%	100% e	75%	78%	77%	50%

a The concentrations of compounds above biphenyl were calculated using DB naphthalene as internal standard, the concentrations of compounds below pyrene using D12 perylene, and the remainder using D10 acenaphthene.

b The less than symbol (<) indicates that the chemical was not detected and that the value is the detection limit.

c Samples were collected in December, 1983 at Eagle Harbor (EGH) and Presidents Point (PPT), Washington.

d Reference to analytical procedure: Malins et al. (1984).

e Deuterated peaks too small to calculate because of dilutions.

Table II.
Concentrations of aromatic compounds in sediment samples, ng/g (ppb) dry weight.

a,b

Site-Station:	EGH-1	EGH-2	EGH-3	EGH-4	EGH-4	EGH-5	EGH-5	EGH-5	EGH-5	EGH-6	EGH-6	EGH-7	EGH-7	EGH-8	EGH-9
1-Propyl Benzene	< 4.0	< 15	< 8.7	< 28	< 28	< 28	37	< 27	< 26	< 20	< 17	< 7.2	< 4.3	< 3.1	< 1.6
n-Propyl Benzene	< 4.3	< 16	< 9.1	1400	1500	< 29	< 28	< 28	< 27	< 21	< 18	< 7.5	< 4.7	< 3.3	< 1.7
Indan	< 4.2	< 15	< 8.7	< 29	< 29	32	< 28	300	390	69	< 18	14	< 4.6	< 3.2	< 1.7
1,2,3,4-Tetraethylbenzene	< 4.3	< 15	< 8.6	< 29	< 30	< 29	< 28	37	47	< 20	< 18	< 7.1	< 4.7	< 3.3	< 1.7
Naphthalene	440	510	160	1000	1000	1300	4300	11000	13000	2100	2100	470	580	170	150
Benzothiophene	< 5.4	38	< 10	< 35	< 35	84	< 36	710	940	130	32	29	< 5.9	< 4.1	< 2.1
2-Methyl Naphthalene	120	160	57	170	360	550	1000	4600	5000	1300	1600	200	220	14	32
1-Methyl Naphthalene	63	71	28	150	110	390	630	2500	2900	970	2000	99	82	11	13
Biphenyl	7.9	64	26	76	89	220	140	1200	1300	490	820	63	24	< 2.4	8.5
2,6-Dimethyl Naphthalene	37	63	25	< 21	30	320	59	1400	1500	610	1100	75	52	< 2.5	9.1
Acenaphthene	110	130	120	290	280	1500	1700	6300	7200	3600	8700	210	210	52	33
2,3,5-Trimethyl Naphthalene	17	< 12	13	< 23	98	120	< 24	490	540	360	980	< 5.1	< 3.3	< 2.5	< 1.4
Fluorene	180	270	150	560	2300	2200	3500	9900	9500	5700	10000	290	510	34	35
Dibenzothiophene	79	140	100	1100	1600	1100	1400	3200	3500	2000	4300	140	140	< 3.1	4.3
Phenanthrene	1100	1100	580	1800	4800	6000	16000	25000	25000	11000	30000	940	1500	170	140
Anthracene	620	790	300	4900	23000	13000	8500	17000	17000	11000	17000	540	2100	130	110
1-Methyl Phenanthrene	140	69	48	1800	2400	330	650	1200	1300	940	2200	50	110	< 2.3	11
3,6-Dimethyl Phenanthrene	50	52	< 5.9	1200	2800	150	80	260	270	250	640	19	14	< 2.4	< 1.3
Fluoranthene	2600	2000	910	71000	74000	12000	17000	16000	19000	15000	28000	1700	1900	410	220
Pyrene	2300	2200	700	48000	50000	11000	15000	22000	27000	12000	16000	1600	1700	360	290
Benz[a]anthracene	1100	1500	540	16000	15000	3600	5500	3300	3700	3100	4700	740	760	130	130
Chrysene	2700	3700	1300	22000	20000	7200	8900	7600	8300	5100	6400	1700	1600	350	250
Benzofluoranthenes	1900	3200	600	920	8900	2900	6600	3400	4000	2000	2400	1400	1400	300	270
Benzofelopyrene	1300	2000	600	5100	4400	1300	4300	1600	1800	930	1300	900	930	260	240
Benzofalpyrene	1000	1600	510	5500	4800	1500	4800	1700	2000	1100	1400	700	750	200	170
Perylene	290	430	120	1200	1000	370	1100	450	510	290	300	160	170	62	37
Indenopyrene	690	820	280	2600	2200	540	4300	540	730	310	< 30	330	370	130	100
Dibenzo[a,h]anthracene	270	330	91	850	720	220	820	260	270	160	< 30	120	120	27	17
Benzofg,h,i]perylene	800	720	190	1500	1200	410	2700	490	560	270	240	280	420	160	170
Sample Weight (g)	10.02	11.38	10.23	10.21	10.13	20.00	10.00	20.00	20.00	20.00	10.23	22.27	10.24	10.04	10.19
% Dry Weight	45.69	46.49	73.24	49.49	49.49	58.76	52.2	60.15	59.29	73.65	72.98	61.87	61.89	65.32	66.12
Recovery of D8 Naphthalene	61%	78%	88%	65%	76%	100% c	100% c	100% c	100% c	100% c	100% c	70%	51%	67%	68%
Recovery of D10 Acenaphthen	72%	78%	86%	70%	81%	100% c	100% c	100% c	100% c	100% c	100% c	73%	60%	73%	72%
Recovery of D12 Perylene	79%	78%	84%	67%	80%	100% c	100% c	100% c	100% c	100% c	100% c	83%	66%	78%	64%
Sum of the AHS	17913.9	21957	7448	189116	222587	68336	109016	142437	137257	80779	142212	12769	15662	2970	2439.9

a Samples were collected in April, 1984 at Eagle Harbor (EGH), Washington.

b Reference to analytical procedure: Malins et al. (1984).

c Deuterated peaks were used to calculate because of dilutions.

Table III. Results of Short-term Bioassays of Eagle Harbor Sediments.

Assay	Test Species	Exposure	Results
I. Amphipod-Sediment Bioassay	<u>Rhepoxynius abronius</u>	10 days to 50 mL sediment/ 900 mL seawater	Eagle Harbor: 100% mortality Dosewallips River Basin: 4% mortality
II. Pacific Oyster Larvae - Sediment Bioassay	<u>Cassostrea gigas</u>	48 hrs to 20 g sediment/1 L seawater	Eagle Harbor: 72% abnormal larvae 88% mortality Dosewallips River Basin: 1% abnormal larvae 2% mortality
III. Bacterial Bioluminescence Assay	<u>Photobacterium phosphoreum</u>	15 min. to organic extracts of sediment	Eagle Harbor: 15 min EC ₅₀ = 0.25 uL/mL Useless Bay: 15 min EC ₅₀ = 7.37 uL/mL
IV. Surf Smelt Larvae - Sediment Bioassay	<u>Hypomesus pretiosus</u>	7 days to suspended particulates prepared by mixing 20 g sediment with 1 L seawater; mixture allowed to stand 1 hr and supernatant collected	Eagle Harbor: 6% solution of particulates caused 100% mortality in 4 days Dosewallips River Basin: 100% solution of particulates caused 16% mortality in 4 days
V. Sand Dollar - Sediment Bioassay	<u>Dendraster excentricus</u>	14 days to 2 L sediment/ 37 L seawater	Eagle Harbor: 100% mortality Useless Bay: 0% mortality
VI. English Sole - Sediment Bioassay	<u>Parophrys vetulus</u>	20 hrs to 37 L sediment/ 230 L seawater	Eagle Harbor: 92% mortality Sand: 0% mortality